IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants:	Alexander P. Moravsky, et al.	**Examiner:**	Ashok Patel
Serial No:	09/680,291	**Art Unit:**	2889
Filed:	October 6, 2000	**Docket:**	21088/14311
For:	DOUBLE-WALLED CARBON NANOTUBES AND METHODS FOR PRODUCTION AND APPLICATION		

Commissioner for Patents
PO Box 1450
Alexandria, Virginia 22313-1450

DECLARATION OF ALEXANDER P. MORAVSKY

I, Alexander P. Moravsky, hereby declare and state as follows:

1. I am one of the co-applicants of the above-identified application, and I have complete knowledge of all aspects of the application.

2. I am currently employed as a Senior Scientist at MER Corporation, Tucson, Arizona, and have been so employed since 1999. My employment history since 1975 is set forth in the Resume of Alexander P. Moravsky, attached hereto as Exhibit 1 and incorporated herein by reference and made a part hereof.

3. I have earned Ph.D, M.Sc. and B.Sc. degrees in the fields of Chemistry, Physics and Molecular and Chemical Physics, all as set forth in my attached Resume in Exhibit 1.

4. I am currently involved in fullerene and carbon nanotube materials synthesis and property studies, and have been so involved since 1991. I have extensive experience and many publications in this field, as set forth in my attached Resume in Exhibit 1.

5. The present application is directed to, *inter alia*, a solid substance comprised by more than one half by weight of hollow carbon nanotubes having walls consisting essentially of two layers of carbon atoms, said nanotubes consisting of two concentric nearly cylindrical graphene layers.

6. I have been advised by counsel that the United States Patent and Trademark Office ("USPTO") has rejected Claim 67 of the present application that claims such a "solid substance comprised by more than one half by weight of hollow carbon nanotubes having walls consisting essentially of two layers of carbon atoms, said nanotubes consisting of two concentric nearly cylindrical graphene layers" as being anticipated by U.S. Patent No. 5,830,326 to Iijima (hereinafter '326 patent). As to the limitation "by more than a half by weight", I have been advised that the Examiner interprets this limitation as "anywhere from 51 percentage of weight up to 100 percentage of weight" in a broadest reasonable manner.

7. In preparing this Declaration, I have reviewed the Office Action dated October 29, 2008 and the '326 patent cited therein.

8. The '326 patent describes a graphite filament which has carbon as a basic structural unit and which has a tubular shape being formed with a helical structure with carbon hexagons that has an outer diameter of 30 nm or less. The tubular lattices, identified in Figure 2 therein as a_1 to a_3, are a multiple structure and the interval between inner and outer tubes is about 0.34 nm.

According to the '326 patent, the graphite filament is prepared as follows: a pair of carbon rod electrodes on which the graphite filaments are grown are placed in the center of a vacuum container, and the air is removed from the container using a vacuum air-removal line

with a vacuum valve attached thereto. When the pressure reaches a prescribed pressure, e.g., about 10^{-6} Torr, the vacuum valve is closed, argon gas is supplied to the container, and the pressure inside the vacuum container is set to a prescribed pressure.

The carbon rod electrodes discussed hereinabove are part of the arc system. One of the electrodes is connected to a positive terminal of an electric arc discharge source and the other electrode is connected to a negative terminal. The positive carbon rod electrode is adjusted using an adjustment device so that there is a suitable gap between the electrodes. The pair of carbon rod electrodes has a diameter of 1cm, and a direct current arc was discharged between the electrodes in argon gas atmosphere (100 Torr). The discharge voltage was 30 V, the discharge current was set to 200 A. According to the '326 patent, on the end of the negative carbon rod electrode, a carbon deposit mass was grown.

As described therein, the deposit was comprised of partially graphitized glass type portion and a graphite filament portion. The graphite filament portion was an accumulation of graphite filaments with an outside diameter of a few nanometers and the length up to about 1 micron.

In another embodiment described in the '326 patent, this process was conducted at 10 Torr. In the product produced at this lower pressure, according to the '326 patent, the portion of graphite filaments was reduced, and the portion of glass type carbon was increased, relative to the product produced at 100 Torr.

9. It is applicants' position that the process in the '326 patent cannot make the DWNTs in the amounts claimed by the present process. This argument was presented in the last Response. More specifically, our arguments consisted of the assessment of the amount of DWNTs that could be produced in the arc system of the '326 patent. This assessment was based

on analysis of the amount of appropriately low diameter carbon nanotubes that have been produced in a similar arc system and under similar process conditions as described in the paper by Kiselev et al., Carbon, v.37, pp. 1093-1103 (1999), a copy of which is attached hereto as Exhibit 18. The assessment revealed an amount of DWNTs, which is about two orders of magnitude below the 50% value claimed in the present application, as recited in Claim 67. However, this argument has not been found persuasive by the Examiner.

To resolve this question of the amount of DWNTs that could be produced in the '326 patent, and thus whether the product claimed in the present application could have been anticipated by the teachings in the '326 patent, the process described in the '326 patent was repeated to determine the amount of DWNTs in the products produced therefrom. The procedure utilized and the results obtained by following the procedure and the arc employed in the '326 patent is described below.

10. All of the acts described herein were performed by me or by scientists and/or technicians working under my direct supervision or control.

11. The arc synthesis experiments were conducted in an experimental setup that comprised a quartz cylindrical chamber with two carbon rod electrodes that have a diameter of 1 cm, and were vertically installed along the chamber axis, in accordance with the setup in the '326 patent. Photographic images of this setup taken before and during the arc operation are shown as Exhibit 2. In all arc runs, the discharge direct current was set to 200 A, and the discharge voltage was 30 V, in accordance with the procedure in the '326 patent. Arc runs were performed in an argon gas atmosphere at 100 Torr pressure (Arc #1) and at 10 Torr pressure (Arc #2), in accordance with description in the '326 patent (See Column 4, lines 46-54 of the '326 patent). During the arc operation, a carbon deposit was grown on the negative carbon-rod electrode

(cathode deposit). A photograph of a cathode deposit is presented in Exhibit 3. The soft black central portion of the cathode deposit contains carbon nanotubes that have been collected from this place and analyzed by High Resolution Transmission Electron Microscopy (HRTEM) and Scanning Electron Microscopy (SEM) techniques.

12. Precise matching of the process conditions to those of the '326 patent ensured obtaining the same products. We were unable to find any differences between the products described in the '326 patent and those generated during reproduction of '326 conditions.

13. A low magnification SEM image of the carbon nanotube containing product from the arc synthesis performed at 100 Torr argon gas pressure (Run "Arc #1") is shown in Exhibit 4. It reveals the presence of a multitude of carbon nanotubes. The outside diameter of these nanotubes has been measured from the SEM images obtained at high magnification. Representative examples of high magnification SEM images are shown in Exhibit 5.

14. HRTEM images were used to determine the number of walls in individual carbon nanotubes and their outside and inside diameters. Representative examples of HRTEM images of individual nanotubes are shown in Exhibits 6-14. The presence of Multi-Walled Carbon Nanotubes (MWNTs) with the number of tube walls ranging from 3 to 52 has been revealed in the whole array of nanotubes observed by HRTEM. Despite a focused search, no Double-Walled Carbon Nanotubes (DWNTs) have been found in HRTEM images, implying that their occurrence is very rare.

15. The number of walls in these MWNTs grew approximately linearly with the outside diameter of the MWNTs, as is shown by the plot presented in Exhibit 15. This is due to the relative constancy of the inside diameter of the MWNTs produced in the arc process under

conditions of runs performed in argon gas atmosphere at 100 Torr (Run "Arc #1") and at 10 Torr (Run "Arc #2"). Therefore, the relative amount of nanotubes with a given number of walls can be determined from the distribution of nanotubes by their outside diameter. This distribution was used to determine the relative amount of DWNTs in the whole array of nanotubes observed with HRTEM and SEM techniques. The outside diameter of every nanotube present in an electron microscopy image has been measured and used in calculating the diameter distribution of the MWNTs.

16. The outside diameter distribution of the MWNTs produced in the runs "Arc #1" and "Arc #2" is presented in Exhibit 16 as a plot of the number of nanotubes against their outside diameter. It is seen from the plot, for example, that the number of nanotubes having an outside diameter in the interval from 13 nm to 14 nm is equal to 43. The distribution peaks at this outside diameter. The nanotubes with this outside diameter are most frequently seen in the electron microscopy images. It is also noted from the plot that there have been only 6 nanotubes observed with outside diameters of less than 7 nm, among the whole array of 479 nanotubes. An outside diameter of 7 nm marks the upper end of the range of stability of DWNTs. It has been observed by M. Motta et al., Advanced Materials, 2007, v.19, pp. 3721-3726, "High Performance Fibres from 'Dog Bone' Carbon Nanotubes" (Exhibit 19) that DWNTs with outside diameters larger than 6 nm are unstable and collapse into a flat 4-layer ribbon structure. So, the number density of DWNTs has an upper limit of about 1% (6 out of 479 tubes). The actual number density of DWNTs is much below this 1% value. The HRTEM micrographs of all 6 of these nanotubes of less than 7 nm diameter are presented in Exhibits 6, 7 and 8. These images show that the number of walls in these nanotubes is equal to or larger than 3. There is much less than one DWNT per 100 MWNTs produced under conditions of the '326 patent.

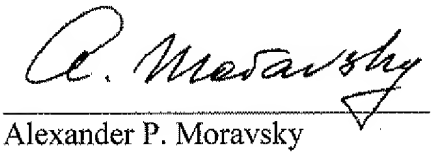
17. It is more common to characterize the content of a component in the multi-component product of a chemical synthesis by weight percentage. Accordingly, the nanotube number distribution discussed above has been transformed into a nanotube mass density distribution over the nanotube outside diameter, which is presented in Exhibit 17. It is seen from this plot that nanotubes with diameters of less than 7 nm comprise about 0.1% of the total mass of all nanotubes. This means that the weight fraction of DWNTs among all the nanotubes produced in the arc synthesis of the '326 patent is much less than 0.1 wt. %, following the considerations given above.

18. Based on the large statistical basis used for determining the amount of DWNTs produced under the conditions of the '326 patent, it is therefore reliably concluded, that the DWNT content in the nanotube product is much less than 0.1 wt.%, while the content by number fraction is much less than 1%. Much less than one DWNT is produced per 100 MWNTs in the arc synthesis of the '326 patent. The process of the '326 patent is unselective towards production of the DWNTs, the selectivity being negligibly small compared to the minimum of 50 wt.% value in Claim 67 of the present application.

19. Thus, it is clearly shown that the teachings in the '326 patent do not anticipate the subject matter claimed in our patent application filed with the USPTO as of October 6, 2000.

20. I further declare that all statements made herein of my knowledge are true and that all statements made on information and belief are believed to be true and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both under Section 1001 of Title 18 of the United States

Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.



Alexander P. Moravsky

November 20, 2009
Date